CORRECTION FACTORS IN GAMMA-RAY DOSIMETRY

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Abstract

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The effect of build-up factors on the measurements of dose have been investigated theoretically and experimentally. The K-fluorescent radiation emission is calculated for cerium and the lighter elements (Cu, Fe and S) to obtain the mass energy transfer coefficients for these elements, and hence the build-up factors of several dose meters – with fluorescent radiation assumed to be (1) all escaping from, and (2) all absorbed in the dose-meter volume. The fractions of the fluorescent radiation that would be absorbed in these dose meters are also estimated. In order to compare the theoretical prediction with experiment, theoretical expressions for the absorbed dose rate and the build-up factors due to an extended source are next derived, which, when expressed in terms of a form factor for the source, retain all the expressions for a point source. Experiments were performed at the Radiation Laboratory of the US Army Natick Research and Development Command in which four chemical dose meters were used: the ferrous-cupric, and the ceric-cerous systems at three different concentrations. The experiments were carried out with a ^{60}Co cylindrical γ-ray source in air and at 64 cm distance from a large ^{60}Co source in a water pool. The build-up factors for these two sources correspond to $\mu r = 0.2$ and 4.25, respectively. It is found experimentally that, for $\mu r = 4.25$, the absorbed dose for a ceric-cerous system with 0.1M ceric-cerous concentration is higher than that with 0.01M ceric-cerous concentration by a factor of 1.25. The theoretical prediction for this factor is 1.21. For four different dose meters, the G-values as determined by these experiments are: 2.33 for 0.01M Ce^{4+} + 0.01M Ce^{3+}, 2.33 for 0.016M Ce^{4+} + 0.009M Ce^{3+}; 1.95 for 0.01M Ce^{4+} + 0.09M Ce^{3+}, all in 0.4M H_2SO_4; and 0.560 for 0.006M $FeSO_4$ + 0.060M $CuSO_4$ in 0.005M H_2SO_4.

1. INTRODUCTION

Absorbed Dose Rate and Buildup Factor

We start with the fundamental equation for the gamma-ray energy absorbed per second in an infinitesimal volume dx dy dz, at a point P(x,y,z). This equation defines the dose rate in $Gy \cdot s^{-1}$ (10^4 $erg \cdot g^{-1} \cdot s^{-1}$),

$$d = 1.60 \times 10^{-10} \int_0^{E\,max} E \cdot \frac{d\phi(E)}{dE} \cdot \frac{\mu_{tr}(E)}{\rho} \cdot dE \qquad (1)$$

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where E = photon energy in MeV

$\phi(E)$ = photon flux density in number of photons per cm^2 per second, i.e., the number of photons with energy less than E which enter a sphere of unit cross sectional area per second at point P

$\frac{\mu_{tr}(E)}{\rho}$ = mass energy transfer coefficient of the material at P for photons of energies lying in the interval between E and E + dE, ρ is the density of the material in $g \cdot cm^{-3}$.

One notes that $E \frac{d\phi(E)}{dE} \cdot dE = dI(E)$

is the photon energy flux density i.e., the intensity, in the energy interval (E, E + dE) in $MeV \cdot cm^{-2} \cdot s^{-1}$. Eq. (1) follows from the basic equation for the absorption of the primary photon flux by $dm = d(\rho x)$ $g \cdot cm^{-2}$ of material,

$$d(dI) = (dI) \cdot \frac{\mu}{\rho} \cdot d(\rho x) = (E d\phi) \cdot \frac{\mu}{\rho} \cdot d(\rho x) \quad (2)$$

when a mass energy transfer coefficient

$\frac{\mu_{tr}(E)}{\rho}$ is used in Eq. (2) instead of the mass total absorption

coefficient (μ/ρ) for the primary photons.

For a Co–60 point source, the energy spectrum at a distance r from the source will consist of two δ functions (the two primary spectral lines) at 1.17 MeV and 1.33 MeV and a continuum background of lower energy photons. The two photons E_1 = 1.17 MeV and E_2 = 1.33 MeV emitted from the source may be approximated, at a distance r, by two photons of E_o = 1.25 MeV each. If the strength of the source is C bequerel, and the attenuation coefficient, or the "total absorption coefficient" of the medium is $\mu(E_o)$ for photons of energy E_o (= 1.25 MeV in the present case), then

$$\frac{dI(E)}{dE} = C \cdot \frac{\exp(-\mu r)}{4\pi r^2} [E_1 \delta(E_1) + E_2 \delta(E_2)] + \text{continuum}$$

$$\triangleq C \cdot \frac{\exp(-\mu r)}{4\pi r^2} \cdot (E_1 + E_2) \cdot \delta\left(\frac{E_1 + E_2}{2}\right) + \text{continuum}$$

The degraded (continuum) part of the spectrum will be denoted by dI(E,r)/dE, and Eq. (1), after integrating out the δ function, becomes

$$d = 1.60 \cdot 10^{-10} \cdot C \cdot \left[\frac{2.5 \exp(-\mu r)}{4\pi r^2} \cdot \frac{\mu_{tr}(E_o)}{\rho} + \int_o^{E_o} \frac{dI(E,r)}{dE} \cdot \frac{\mu_{tr}(E)}{\rho} \cdot dE\right]$$

$$\text{or } d = 1.60 \cdot 10^{-10} \cdot C \cdot I_o \frac{\exp(-\mu r)}{4\pi r^2} \cdot \frac{\mu_{tr}(E_o)}{\rho} \cdot \left[1 + \frac{\int_o^{E_o} \exp(\mu r) \cdot 4\pi r^2 \frac{dI(E,r)}{dE} \cdot \frac{\mu_{tr}(E)}{\rho} dE}{I_o \frac{\mu_{tr}(E_o)}{\rho}} \right] \quad (3)$$

where $I_o = 2 \cdot 1.25$ MeV

$E_o = 1.25$ MeV

Had there been no degradation of the spectrum, we would have $dI(E,r)/dE = 0$ and Eq. (3) would become

$$d = 1.60 \cdot 10^{-10} \cdot C \cdot I_o \frac{\exp(-\mu r)}{4\pi r^2} \cdot \mu_{tr}(E_o)/\rho \quad (4)$$

i.e., the expression in the bracket of Eq. (3)

$$B(r) \equiv 1 + \frac{\int_o^{E_o} \exp(\mu r) \cdot 4\pi r^2 \cdot \frac{dI(E,r)}{dE} \cdot \frac{\mu_{tr}(E)}{\rho} dE}{I_o \frac{\mu_{tr}(E_o)}{\rho}} \quad (5)$$

becomes unity.

The primary line spectrum is always degraded into a line spectrum (of smaller intensity) given by the factor $(I_o \exp(-\mu r)/4\pi r^2)$ and a continuum part due to interactions, the integral term inside the brackets of Eq. (3), which thus accounts for a genuine gamma-ray buildup in the medium as the primary photons propagate through it.

B(r) of Eq. (5) is called, therefore, the buildup factor for the absorbed dose rate or the absorbed dose at point P(x,y,z).

Eq. (3) may then be written as

$$d = 1.60 \cdot 10^{-10} C \cdot I_o \cdot \frac{\exp(-\mu r)}{4\pi r^2} \cdot \frac{\mu_{tr}(E_o)}{\rho} \cdot B(r) \quad (6)$$

If at point P, we place a dosimeter, the rate of the dose absorbed by it will be given by Eq. (6). The buildup factor B is given by Eq. (5). From these equations it is seen that, in order to arrive at an accurate value of the absorbed dose rate, it is important that we know accurately the spectral distribution $dI(E,r)/dE$ of the degraded gamma rays at P(x,y,z), and the mass energy transfer coefficient $\mu_{tr}(E)/\rho$ of the materials which make up the dosimeter, for the entire energy range covered.

2. ENERGY TRANSFER COEFFICIENT WITH FLUORESCENT RADIATION CORRECTION 1

The mass energy transfer coefficient is the sum of the three absorption coefficients derived from the interaction cross-sections for photoelectric absorption, Compton scattering and pair production, and corrected for the energy non-absorptive parts of the three interactions:

$$\frac{\mu_{tr}(E)}{\rho} = \frac{\tau}{\rho}\left(1-\frac{\delta}{h\nu}\right) + \frac{\sigma_a}{\rho} + \frac{\kappa}{\rho}\left(1-\frac{2mc^2}{h\nu}\right) \tag{7}$$

where τ/ρ = photoelectric mass absorption coefficient

σ_a/ρ = the energy absorption component of the Compton process

κ/ρ = mass absorption coefficient due to pair production

$1-\delta/(h\nu)$ = correction that accounts for the fraction of energy escaping as fluorescent radiation per photoelectric interaction at energy $E = h\nu$

$1-2mc^2/(h\nu)$ = rest mass correction factor for the pair production to account for the escaping annihilation photons.

The mass energy transfer coefficient of Eq. (7) is to be distinguished from the so-called "mass absorption coefficient"

$$\frac{\mu_a}{\rho} = \frac{\tau}{\rho} + \frac{\sigma_a}{\rho} + \frac{\kappa}{\rho} \tag{8}$$

The cross sections for the three fundamental processes are well known so that accurate values of μ_{tr}/ρ can be obtained when correction for fluorescent radiation $1 - \delta/h\nu$ is known. For a Co–60 source, the correction for the pair production term, $(1-2mc^2/h\nu)$, can be neglected.

In Reference [1], it was assumed that the fluorescent radiation emitted is absorbed again by the dosimeter, i.e. $\delta=0$.

In most dosimeters applied to measure dose in biological systems this is a good assumption. But if the dosimeter contains high atomic number elements such as Ce, a significant amount of the fluorescent radiation may often escape.

We have extended, therefore, the calculations in Reference [1] by: (I) assuming that the fluorescent radiation entirely escapes from the dosimeter, (II) estimating the part of the fluorescent radiation that would be absorbed again by the dosimeter.

For correction I, we calculate, for each energy, the first term of Eq. (7), the photoelectric absorption corrected for fluorescent radiation emission, according to the following equation

$$\frac{\tau}{\rho}\left(1-\frac{\delta}{h\nu}\right) = \frac{\tau}{\rho}\left(1-\frac{\omega_K \cdot E_K}{h\nu}\right) \qquad (9)$$

where ω_K is the fluorescent yield,

$$\omega_K = \frac{\text{number of K-shell X-rays}}{\text{number of K-shell X-rays + number of Auger electron events}}$$

and E_K = K=shell energy

In Eq. (9), we consider only the fluorescent radiation from the K-shell, since fluorescent radiation from the L and higher shells, being very much softer, will be practically all absorbed in the volume under consideration.

The fluorescent emission does not change the photoelectric cross section, hence it does not change the photoelectric mass absorption coefficient τ/ρ, but it does make the energy transfer E that is locally absorbed smaller by a factor of $(1 - \delta/E)$. Inspection of Eqs. (1) and (2) shows that it is convenient to incorporate this factor with the absorption coefficient μ_a/ρ, and re-define this new coefficient as the mass energy transfer coefficient.

The same remark holds for the correction factors for the other two terms in Eq. (7) for Compton scattering and pair production.

Table I gives for cerium, the values of τ/ρ and μ_{tr}/ρ thus calculated, and $(\mu_{tr}/\rho)_{FA}$, the mass energy transfer coefficient with fluorescent radiation assumed to be all absorbed (FA) again in the volume under consideration. It will be noted that the values of $(\mu_{tr}/\rho)_{FA}$ are just that of μ_a/ρ. The values of τ/ρ and $(\mu_{tr}/\rho)_{FA}$ are from the compilation by Storm, Gilbert and Israel [2], with interpolation and extrapolation for the values at those energies not given by these authors.

The values of these three coefficients, τ/ρ, μ_{tr}/ρ and $(\mu_{tr}/\rho)_{FA}$, for Cu, Fe and S are similarly computed and are given in Tables II, III, and IV.

In Figures 1 and 2, we show τ/ρ and μ_{tr}/ρ for Ce, Fe and S.

In calculating the correction for fluorescent emission, Eq. (9), the fluorescent yield ω_K for the K-shell is computed from the following equation given by Hagedoorn and Wapstra [3].

$$\frac{\omega_K}{1-\omega_K} = (-6.4 \cdot 10^{-2} + 3.4 \cdot 10^{-2}\ Z - 1.03 \cdot 10^{-6} Z^3)^4 \qquad (10)$$

where Z = atomic number. For Ce, Cu, Fe, and S, it is equal to 0.89, 0.39, 0.29, and 0.049, respectively.

3. CALCULATIONS OF THE BUILDUP FACTORS WITH FLUORESCENT RADIATION CORRECTION I

To calculate the absorbed dose rate d and the buildup factor B in Eqs. (5) and (6), we need the values of the spectral distribution $dI(E,r)/dE$ at point P. We use, as

TABLE I*

τ/ρ, μ_{tr}/ρ AND (μ_{tr}/ρ) FOR CERIUM

Energy (MeV)	τ/ρ ($cm^2 \cdot g^{-1}$)	μ_{tr}/ρ ($cm^2 \cdot g^{-1}$)	$(\mu_{tr}/\rho)_{FA}$ ($cm^2 \cdot g^{-1}$)
0.01	188.0	188.0	188.0
0.015	58.5	58.5	58.6
0.02	25.9	25.9	25.9
0.03	7.92	7.92	7.92
0.04	3.45	3.45	3.45
0.040449	3.22	3.20	3.22
	25.44	2.794	25.44
0.05	14.20	3.977	14.20
0.06	8.85	3.56	8.87
0.08	4.10	2.26	4.11
0.10	2.20	1.42	2.21
0.15	0.688	0.543	0.708
0.20	0.306	0.274	0.329
0.30	0.103	0.115	0.127
0.40	0.0456	0.0659	0.070
0.50	0.0250	0.0476	0.0494
0.60	0.0160	0.0397	0.0406
0.80	0.0084	0.0319	0.0323
1.00	0.0055	0.0285	0.0287
1.25	0.0040	0.0268	0.0268
1.50	0.0027	0.0247	0.0247

*τ/ρ = mass photoelectric absorption coefficient

μ_{tr}/ρ = mass energy transfer coefficient with fluorescent ratiation assumed to be all escaping from the volume under consideration

$(\mu_{tr}/\rho)_{FA}$ = mass energy transfer coefficient with fluorescent radiation assumed to be all absorbed again in the volume under consideration

in reference [1], the values of exp $(\mu r) \cdot 4\pi r^2 \cdot dI(E,r)/(dE)$ given by Goldstein and Wilkins [4] for a point isotropic Co—60 source immersed in water, at distances of μr = 1, 2, and 4.

Table V gives the buildup factors B_{FE} and B_{FA} so calculated. B_{FE} denotes the case when the fluorescent radiation is assumed to escape entirely from the volume under consideration and B_{FA} the case when all the fluorescent radiation is absorbed.

TABLE II*

τ/ρ, μ_{tr}/ρ AND $(\mu_{tr}/\rho)_{FA}$ FOR COPPER

Energy (MeV)	τ/ρ ($cm^2 \cdot g^{-1}$)	μ_{tr}/ρ ($cm^2 \cdot g^{-1}$)	$(\mu_{tr}/\rho)_{FA}$ ($cm^2 \cdot g^{-1}$)
0.01	224.0	145.6	224.0
0.015	75.8	58.1	75.8
0.02	33.9	27.97	33.9
0.03	10.6	9.36	10.6
0.04	4.49	4.11	4.50
0.05	2.29	2.15	2.31
0.06	1.35	1.29	1.37
0.08	0.570	0.562	0.587
0.10	0.291	0.300	0.310
0.15	0.084	0.105	0.107
0.20	0.035	0.0588	0.0594
0.30	0.010	0.0366	0.0367
0.40	0.0045	0.0315	0.0315
0.50	0.0025	0.0296	0.0296
0.60	0.0016	0.0286	0.0286
0.80	0.0008	0.0271	0.0271
1.00	0.0005	0.0260	0.0260
1.25	0.0003	0.0249	0.0249
1.50	0.0002	0.0240	0.0240

*See footnote of Table I for the meaning of the three coefficients.

The values of B_{FA} given in Table V differ somewhat from those given in Table II of the previous calculations [1]; e.g. for Ce, it is 236 instead of 241. This is because different widths ΔE were used in the numerical integration of the integral in Eq. (5), and the interpolated and extrapolated values of μ_{tr}/ρ and $\exp(\mu r) \cdot 4\pi r^2 \cdot dI(E,r)/(dr)$ were read from the graphs in both cases. It should be emphasized, however, that we are here calculating the corrections. A difference of a few percent in the corrections will not affect the results and the conclusions in any way.

4. FLUORESCENT RADIATION CORRECTION II

In the above calculations, we assume that the fluorescent radiation emitted from the dosimeter materials either all escapes from the dosimeter, giving thus the buildup factor B_{FE}, or is entirely absorbed by the dosimeter, giving thus the buildup factor B_{FA}. The actual situation, of course, will lie between these two extreme cases, i.e., the fluorescent

TABLE III*

τ/ρ, μ_{tr}/ρ AND $(\mu_{tr}/\rho)_{FA}$ FOR IRON

Energy (MeV)	τ/ρ ($cm^2 \cdot g^{-1}$)	μ_{tr}/ρ ($cm^2 \cdot g^{-1}$)	$(\mu_{tr}/\rho)_{FA}$ ($cm^2 \cdot g^{-1}$)
0.01	178.0	141.3	178.0
0.015	58.0	50.03	58.0
0.02	25.7	23.05	25.7
0.03	7.88	7.35	7.89
0.04	3.32	3.16	3.33
0.05	1.67	1.61	1.68
0.06	0.981	0.961	0.995
0.08	0.410	0.416	0.427
0.10	0.206	0.221	0.225
0.15	0.058	0.0801	0.0809
0.20	0.024	0.0482	0.0484
0.30	0.0071	0.0340	0.0340
0.40	0.0031	0.0306	0.0306
0.50	0.0017	0.0293	0.0293
0.60	0.0011	0.0287	0.0287
0.80	0.0005	0.0274	0.0274
1.00	0.0003	0.0264	0.0264
1.25	0.0002	0.0252	0.0252
1.50	0.0002	0.0243	0.0243

*See footnote of Table I for the meaning of the three coefficients.

radiation emitted is only partially absorbed by the dosimeter, the unabsorbed part will escape from the dosimeter to the surrounding medium.

The portion of the fluorescent radiation absorbed by the dosimeter could be estimated as follows.

If the photon flux density is $d\phi(E)$, then the induced fluorescent photon flux is $\omega_K \cdot d\phi(E) \cdot \tau_{h\nu}/\rho$ per $g \cdot cm^{-2}$ thickness of the fluorescent radiation-producing material where $h\nu=E$ is the energy of the primary photons. If E_K is the photon energy in the fluorescent radiation, and $\mu_{tr}(E_K)/\rho$ the mass energy transfer coefficient of the material

TABLE IV*

τ/ρ, μ_{tr}/ρ AND $(\mu_{tr}/\rho)_{FA}$ FOR SULPHUR

Energy (MeV)	τ/ρ ($cm^2 \cdot g^{-1}$)	μ_{tr}/ρ ($cm^2 \cdot g^{-1}$)	$(\mu_{tr}/\rho)_{FA}$ ($cm^2 \cdot g^{-1}$)
0.01	50.5	49.9	50.5
0.015	15.5	15.4	15.5
0.02	6.5	5.46	5.5
0.03	1.88	1.88	1.89
0.04	0.744	0.753	0.755
0.05	0.353	0.366	0.367
0.06	0.200	0.216	0.216
0.08	0.080	0.0980	0.0981
0.10	0.039	0.0596	0.0596
0.15	0.010	0.0348	0.0348
0.20	0.0043	0.0310	0.0310
0.30	0.0010	0.0301	0.0301
0.40	0.0006	0.0301	0.0301
0.50	0.0003	0.0300	0.0300
0.60	0.0002	0.0298	0.0298
0.80	0.0001	0.0289	0.0289
1.00	0.00006	0.0280	0.0280
1.25	0.00004	0.0265	0.0265
1.50	0.00000	0.0257	0.0257

*See footnote of Table I for the meaning of the three coefficients.

TABLE V

ABSORBED DOSE BUILDUP FACTORS B_{FE} AND B_{FA} FOR S, Fe, Cu AND Ce AT DIFFERENT DISTANCES FROM A POINT ISOTROPIC CO–60 SOURCE IMMERSED IN WATER*

Element	Z	Distance					
		$\mu r = 1$		$\mu r = 2$		$\mu r = 4$	
		B_{FE}	B_{FA}	B_{FE}	B_{FA}	B_{FE}	B_{FA}
S	16	3.43	3.44	6.86	6.87	13.9	13.9
Fe	26	8.60	9.00	20.2	21.2	43.6	45.7
Cu	29	10.9	11.8	26.1	28.3	56.7	61.5
Ce	58	19.0	38.4	47.1	102.0	106.7	236.1

*Subscript FE denotes that all of the fluorescent radiation escapes and FA denotes that all of the fluorescent radiation is absorbed.

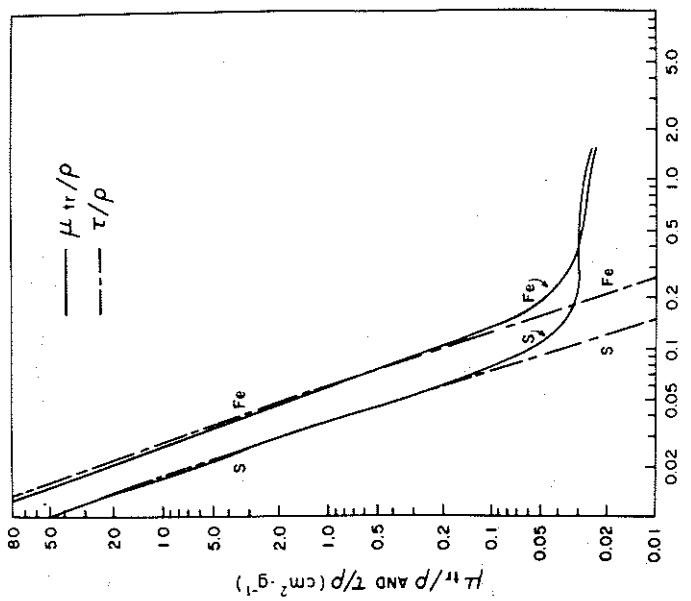

FIG.2. Mass energy transfer coefficient and photoelectric mass absorption coefficient for iron and sulphur.

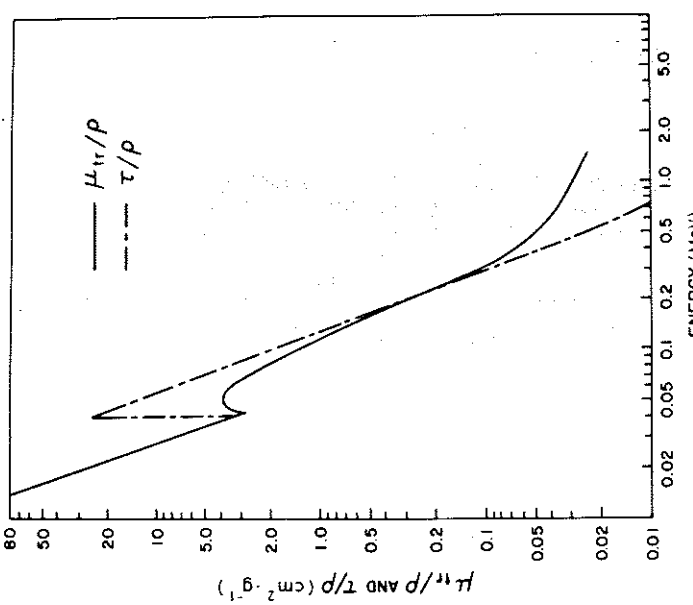

FIG.1. Mass energy transfer coefficient and photoelectric mass absorption coefficient for cerium.

at energy E_k, then the fluorescent energy absorbed by an effective thickness $\Delta m = \Delta(\rho \ell)$ of the material is equal to

$$\text{(Fluorescent energy flux density)} \cdot (\mu_{tr}(E_k)/\rho) \cdot \Delta m$$

$$= (\omega_k \cdot d\phi(E) \cdot E_k \cdot \frac{\tau_{h\nu}}{\rho}) \cdot \frac{\mu_{tr}(E_k)}{\rho} \cdot \Delta(\rho \ell) \qquad (11)$$

for small values of $(\mu_{tr}(E_k)/\rho) \cdot \Delta m$.

For a ceric dosimeter of 0.1M $Ce(SO_4)_2$ and 0.4M H_2SO_4, the effective $[\mu_{tr}(E_k)/\rho] \cdot \Delta(\rho \ell)$ for $\ell = 1$ cm, is

$$\left[\frac{\mu_{tr}(E_k)}{\rho} \cdot \Delta(\rho \ell)\right]_{\text{effective}}$$

$$= \left[\left(\frac{\mu_{tr}}{\rho}\right)_{H_2O} \cdot 0.994 + \left(\frac{\mu_{tr}}{\rho}\right)_{Ce} \cdot \frac{0.1\, A_{Ce}}{1000} + \left[\left(\frac{\mu_{tr}}{\rho}\right)_{S} \cdot \frac{32}{96} + \left(\frac{\mu_{tr}}{\rho}\right)_{O} \cdot \frac{64}{96} \right] \cdot \frac{0.6\, A_{SO_4}}{1000} + \left(\frac{\mu_{tr}}{\rho}\right)_{H} \cdot \frac{0.8 \cdot A_H}{1000} \right] \cdot 1.066$$

$$= 0.131 \text{ (for } \ell = 1 \text{ cm)} \qquad (12)$$

$$\text{or} = 0.123 \text{ (for } \Delta(\rho \ell) = 1 \text{ g cm}^{-2}) \qquad (13)$$

Thus for such a ceric dosimeter, per centimetre dimension, about 13% of the K-fluorescent energy from ceric atoms will be re-absorbed by the dosimeter.

From Berger's calculation [5], we see that if the dosimeter was water-equivalent, approximately 7.7% of the fluorescent radiation would be absorbed per cm of the dosimeter. The ceric dosimeter solution of 0.1M has $\mu_{tr}/\rho = 0.123$ versus $\mu_{tr}/\rho = 0.062$ for water, we therefore multiply 7.7% by 0.123/0.062 = 15%, in good agreement with our estimation, Eq. (12), given above for the percentage of absorption of the fluorescent radiation per cm in such a ceric dosimeter.

For a ceric dosimeter of 0.01M $Ce(SO_4)_2$ and 0.4M H_2SO_4, the corresponding percentage of absorption of the K-fluorescent radiation by the dosimeter is about 7% per cm dimension of the dosimeter.

5. BUILDUP FACTOR FOR A COMPOSITE MATERIAL

We now calculate the buildup factors for ceric dosimeters of two different ceric concentrations, for three different distances from the point isotropic Co–60 source immersed in water, since such calculations with fluorescent radiation assumed to be totally absorbed show strong ceric concentration dependence [1].

TABLE VI

BUILDUP FACTORS $B(r)_{FE}$ AND $B(r)_{FA}$ FOR TWO CERIC DOSIMETERS AT DIFFERENT DISTANCES FROM A POINT ISOTROPIC CO–60 SOURCE IMMERSED IN WATER

Dosimeter	Distance					
	$\mu r = 1$		$\mu r = 2$		$\mu r = 4$	
	B_{FE}	B_{FA}	B_{FE}	B_{FA}	B_{FE}	B_{FA}
Dosimeter (1) 0.01M $Ce(SO_4)_2$ 0.4M H_2SO_4 H_2O	2.071	2.095	3.407	3.475	6.326	6.485
Dosimeter (2) 0.1M $Ce(SO_4)_2$ 0.4M H_2SO_4 H_2O	2.258	2.490	3.891	4.546	7.420	8.975
Ratio (2)/(1)	1.090	1.189	1.142	1.308	1.173	1.384

TABLE VII

ABSORBED DOSES RELATIVE TO WATER, d_s/d_w, OF THE TWO CERIC DOSIMETERS AT DIFFERENT DISTANCES FROM A POINT ISOTROPIC CO–60 SOURCE IMMERSED IN WATER

Dosimeter	Distance					
	$\mu r = 1$		$\mu r = 2$		$\mu r = 4$	
	$(d_s/d_w)_{FE}$	$(d_s/d_w)_{FA}$	$(d_s/d_w)_{FE}$	$(d_s/d_w)_{FA}$	$(d_s/d_w)_{FE}$	$(d_s/d_w)_{FA}$
Dosimeter (1) 0.01M $Ce(SO_4)_2$ 0.4M H_2SO_4 H_2O	1.015	1.027	1.025	1.046	1.032	1.058
Dosimeter (2) 0.1M $CE(SO_4)_2$ 0.4M H_2SO_4 H_2O	1.103	1.217	1.167	1.364	1.207	1.460

From Eq. (5) for the buildup factor, it follows that the buildup factor for a composite material made up of elements X_1, X_2, X_3 .... with the buildup factors B_1, B_2, B_3 .... and mass energy transfer coefficients μ_1/ρ_1, μ_2/ρ_2, μ_3/ρ_3 .... at 1.25 MeV photon energy, and weight fractions A_1, A_2, A_3 ...., is given by Eq. (12) of [1],

$$B(r) = \frac{B_1(r) \cdot \frac{\mu_1}{\rho_1} \cdot A_1 + B_2(r) \cdot \frac{\mu_2}{\rho} \cdot A_2 + \ldots}{\frac{\mu_1}{\rho_1} \cdot A_1 + \frac{\mu_2}{\rho_2} \cdot A_2 + \ldots} \qquad (14)$$

since the mean mass energy transfer coefficient of the composite material is

$$\overline{\left(\frac{\mu}{\rho}\right)} = \frac{\mu_1}{\rho_1} \cdot A_1 + \frac{\mu_2}{\rho_2} \cdot A_2 + \ldots \qquad (15)$$

The buildup factors B_{FE} and B_{FA} for the two ceric dosimeters thus calculated are given in Table VI, where we also show the ratios of the corresponding buildup factors for the two dosimeters.

In Table VII we give the ratio of the absorbed dose rates d_s in these two dosimeters relative to the absorbed dose rate d_w in water,

$$\frac{d_s}{d_w} = \frac{(\mu_{tr}/\rho)_s \cdot B_s(r)}{(\mu_{tr}/\rho)_w \cdot B_w(r)} \qquad (16)$$

using the value of $(\mu_{tr}/\rho)_w \cdot B_w(r)$ previously calculated for water.

6. COMPARISONS OF THEORY WITH EXPERIMENT

A series of experiments was performed from February to July 1976 in order to demonstrate the rather large differences in the dose absorbed by dosimeters of different materials, and which have been irradiated with the same source for the same duration. We report here the results obtained with a Fe–Cu dosimeter and three ceric dosimeters of different ceric-cerous concentrations.

6.1 Dosimeters Used

The Fe–Cu dosimeter solution has the following composition – 0.006M $FeSO_4$, 0.060M $CuSO_4$ and 0.005M H_2SO_4, and that of the three ceric dosimeters: 0.01M Ce^{4+} and 0.01M Ce^{3+}, 0.016M Ce^{4+} and 0.009M Ce^{3+}, and 0.01M Ce^{4+} and 0.09M Ce^{3+}, all in 0.4M H_2SO_4.

The ampules used were 3.7 x 1.63 cm diameter.

These four dosimeters will be referred to as FeCu (6/60), Ce (10/10), Ce (16/9) and Ce (10/90). Their buildup factors B(r) are calculated as before for the two cases, a) all fluorescent radiation escapes (FE) and, b) all fluorescent radiation is absorbed (FA). The values are tabulated in Table VIII along with their density ρ and their $\mu_{tr}(E_0)/\rho$.

TABLE VIII

BUILDUP FACTORS OF THE FOUR DOSIMETERS USED

Dosimeter	ρ	$\frac{\mu_{tr}(E_o)}{\rho}$	Distance						
			$\mu r = 1$		$\mu r = 2$		$\mu r = 4$		
			$B(r)_{FE}$	$B(r)_{FA}$	$B(r)_{FE}$	$B(r)_{FA}$	$B(r)_{FE}$	$B(r)_{FE,\ partial}$	$B(r)_{FA}$
Ce(10/10)	1.0489	0.02964	2.092	2.139	3.462	2.600	6.448	6.504	6.761
Ce(16/9)	1.0517	0.02965	2.101	2.159	3.485	3.651	6.502	6.572	6.892
Ce(10/90)	1.0936	0.02958	2.252	2.477	3.874	4.512	7.394	7.794	8.874
FeCu(6/60)	1.011	0.02971	(2.061)	2.061	(3.391)	3.391	6.293	6.295	6.308

6.2 Gamma-Ray Sources Used

Two gamma-ray sources were used: a) the NUMEC-source which contains $6 \cdot 10^{14}$ Bq and consists of about 25 cm long Co–60 strips arranged in a cylindrical configuration of 7.6 cm radius, and, b) the main Co–60 source which contains $8.1 \cdot 10^{16}$ Bq and consists of two parallel plaques of Co–60 strips. The two plaques are 146 cm x 180.3 cm in area and 53.3 cm apart. The dosimeters were placed on a perpendicular to the center of the plaques and 64 cm away from the nearest plaque.

In the NUMEC, the value of μr is equivalent to about 3 cm of water, or a $\mu r \triangleq 0.2$, which accounts for the effect of the source thickness and the ampule size. The main source was under water and in the dosimeter position the effective value of $\mu r = 4.25$.

6.3 Spectrophotometer Used

A Cary 15 spectrophotometer was used for measurement of the optical density changes of the dosimeter solutions in the usual way for determining the changes in the ceric concentrations.

6.4 Generalized Expression of the Absorbed Dose Rate Due to an Extended Source

It can be shown that for an actual extended source, the absorbed dose rate at point P(x,y,z) is still given by a generalized equation of the form of Eq. (6):

$$d = 1.60 \times 10^{-10} \cdot I(r) \cdot \frac{\mu_{tr}(E_o)}{\rho} \cdot B(r) \qquad (17)$$

where

$$I(r) = \Sigma_i I(r_i) = \Sigma_i C_i \cdot I_o \cdot \frac{\exp(-\mu r_i)}{4\pi r_i^2} \qquad (18)$$

$$B(r) = \frac{\Sigma_i I(r_i) \cdot B(r_i)}{\Sigma_i I(r_i)} \qquad (19)$$

r_i = distance between source element i and point P(x,y,z) = P $(\vec{r})$.

For a continuous distribution, Σ_i is to be replaced by integrals.

B(r) in Eq. (19) for an extended source may be approximated by the buildup factor for a point source at some arbitrarily chosen point r_o and multiplied by a "form factor" $F(r,r_o)$ of the extended source such that:

$$B(r)|_{\text{extended source}} = B(r) = B(r_o)_{\text{point source}} \cdot F(r,r_o) \qquad (10a)$$

We note that the form factor $F(r,r_o)$ for a finite source will approach unity at great distances from the source

$$F(r,r_o) \rightarrow 1, \qquad \text{for } r,r_o \rightarrow \infty \qquad (19b)$$

TABLE IX

SUMMARY OF EXPERIMENTAL RESULTS

Irradiator and Medium	Dosimeter	ΔOD Observed Mean	G(X)*	ΔOD(Ce)/ΔOD(FeCu(6/60)) Expt	ΔOD(Ce)/ΔOD(FeCu(6/60)) Theory	ΔOD(Ce)/ΔOD(Ce(10/10)) Expt	ΔOD(Ce)/ΔOD(Ce(10/10)) Theory
NUMEC (Air)	Ce(10/10)	52.30	2.33	10.95	10.95	1.000	1.000
	Ce(16/9)	52.47	2.33	10.99	10.99	1.003	1.003
	Ce(10/90)	45.53	1.95	9.54	9.54	0.871	0.871
	FeCu(6/60)	4.775	0.560				
MAIN UNDERWATER (Water)	Ce(10/10)	34.18	(2.33)	11.32	11.28		
	FeCu(6/69)	3.018	(0.560)				
	Ce(16/9)	56.18	(2.33)	11.59	11.43	1.00	1.000
	FeCu(6/60)	4.848	(0.560)				
	Ce(10/90)	36.80	(1.95)	12.19	11.65	1.08	1.044
	FeCu(6/69)	3.018	(0.560)				
	Ce(10/90)	36.95	(1.95)			1.10	1.044
	Ce(10/10)	33.48	(2.33)				

* Based on molar extinction coefficient of Ce^{3+} at 320 nm = 5580 $M^{-1}cm^{-1}$ and of Fe^{3+} at 305 nm = 2200 $M^{-1}cm^{-1}$.

6.5 Method of Analysis

The absorbed dose D measured by a chemical dosimeter is proportional to the change of optical density ΔOD

$$D = K' \frac{\Delta OD}{\epsilon \cdot G(X) \cdot \rho \cdot \ell} \tag{20}$$

where $K' = 0.9647 \times 10^7$ when D is in grays, and other symbols have their usual meanings, ϵ = the molar extinction coefficient, G(X) the molecular changes per 100 eV, ρ the density, and ℓ the dimension of the photometric cell.

Thus, with the same exposure, two dosimeters will have the ratio of their ΔOD's as follows:

$$\frac{(\Delta OD)_1}{(\Delta OD)_2} = \frac{\epsilon_1 \cdot G(X_1) \cdot \rho_1}{\epsilon_2 \cdot G(X_2) \cdot \rho_2} \cdot \frac{D_1}{D_2} \tag{21}$$

and D_1 and D_2 are given by Eq. (17) and the irradiation time t

$$D_1 = d_1 \cdot t \tag{22}$$

$$D_2 = d_2 \cdot t$$

so that

$$\frac{(\Delta OD)_1}{(\Delta OD)_2} = \frac{\epsilon_1 \cdot G(X_1) \cdot \rho_1}{\epsilon_2 \cdot G(X_2) \cdot \rho_2} \cdot \frac{(\mu_{tr}/\rho)_1}{(\mu_{tr}/\rho)_2} \cdot \frac{B_1(r)}{B_2(r)} \tag{23}$$

6.6 Experimental Results

Table IX summarizes the results obtained. The mean observed ΔOD's listed in the Table were each obtained from a number of experiments. They are corrected for decay and normalized to the same irradiation time. The error in each experimental determination is estimated to be about 2%. For the experiments in the NUMEC irradiator, accurate doses of the experiments were available from separate determinations using Fe--Cu dosimeters which had been calibrated against the Fricke dosimeter. From the observed ΔOD and for a given dose, G(X) values of the four dosimeters were calculated using Eq. (20) for $\epsilon_{Fe} = 2200$ and $\epsilon_{Ce} = 5580$. The values are listed in the Table IX.

From the observed ΔOD, we obtained the experimental values of the ratios of ΔOD,

$$\frac{\Delta OD\ (Ce)}{\Delta OD\ (FeCu\ (6/60))} \quad \text{and} \quad \frac{\Delta OD\ (Ce)}{\Delta OD\ (Ce(10/10))}$$

which are shown in Table IX along with the theoretical predictions from Eq. (23) with the B(r) values calculated for the case of partial escape of the fluorescent radiation and listed in Table VIII as $B(r)_{FE,\,partial}$ for $\mu r = 4$ for the UNDERWATER experiments.

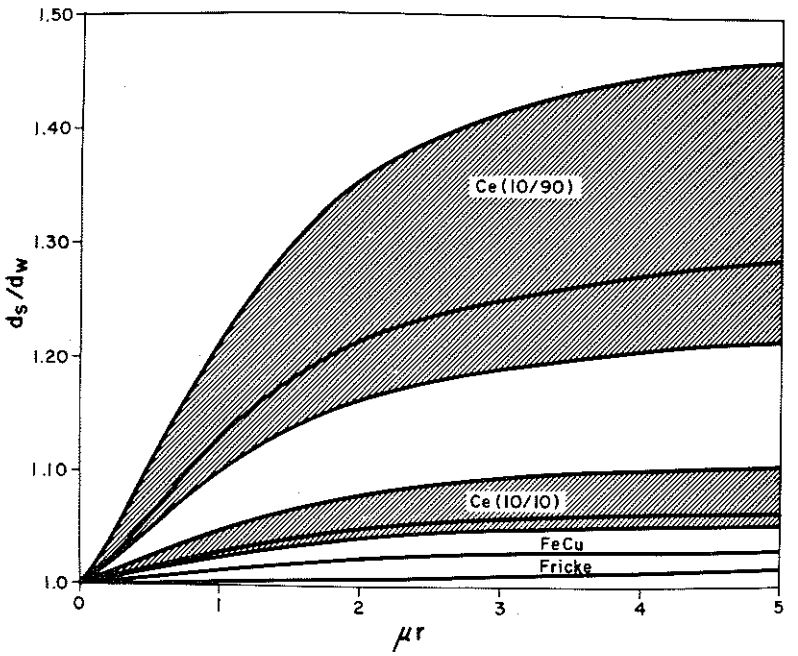

FIG.3. d_s/d_w, the absorbed dose rate of dosimeter relative to that of water, as a function of distance, for different dosimeters: Ce(10/90), Ce(10/10), FeCu(6/60) and the Fricke. The partial re-absorption of the K-fluorescent radiation in the dosimeter gives bands (the shaded areas) for the ceric-cerous systems. The two curves within the bands are for dosimeters with ampules of 3.7 cm X 1.63 cm diameter.

In Figure 3, we show the ratio d_s/d_w of the absorbed dose rate (d_s) of dosimeter Ce(10/90), Ce(10/10), FeCu(6/60) and the Fricke, to that of water (d_w), as a function of distance μr. For Ce(10/90) and Ce(10/10), the partial re-absorption of the K-fluorescent radiation in the dosimeter ampule that is 3.7 cm high and 1.63 cm in diameter, gives the curves in the two shaded bands.

It is estimated that for the dimension of the dosimeters used, the percentage of absorption of the fluorescent radiation is about 18% for Ce(10/10) and Ce(16/9), and about 27% for Ce(10/90).

In the NUMEC irradiator the ΔOD values obtained are used to calculate the G-values. But the exact check on the theory can first be made when we compare these values with the determination of ΔOD for dosimeters at some distance under water from the source. It is seen from Table IX that the agreement between theory and experiment is satisfactory.

But the effects of the buildup factors going into the absorbed dose formula for a dosimeter can be seen in an even more striking way when we form the "water" to "air" ratio of the optical density changes ΔOD, i.e.:

$\frac{\Delta OD(Ce(10/90))}{\Delta OD(Ce(10/10))}$. In this "water" to "air" ratio, all factors of Eq. (23) cancel out except the four B(r)'s for water and air.

$$\frac{\left[\frac{\Delta OD(Ce(10/90))}{\Delta OD(Ce(10/10))}\right]_{water}}{\left[\frac{\Delta OD(Ce(10/90))}{\Delta OD(Ce(10/10))}\right]_{air}} = \frac{\frac{1}{2}(1.08 + 1.10)}{0.87} = \frac{1.09}{0.87} = 1.25 \text{ (Experimental)} \quad (24)$$

with an uncertainty in the numerator and in the denominator of about 2%, or in the overall "water" to "air" ratio of about 4%.

The theoretical value of the ratio of Eq. (24)

$$\text{Ratio on L.H.S. of Eq. (24)} = \frac{1.044}{0.871} \times \text{(correction factor for extended source)}$$

$$= 1.21 \quad \text{(Theoretical)}$$

where the correction factor for the source (a plane source versus point source) is about 1.01. Had we used Berger's value [5] for the absorption of soft gamma rays comparable to the fluorscent radiation rather than our rough estimate in Eq. (12), we would have obtained a value of 1.24 to be compared with the experimental value of 1.25.

The agreement between the theoretical and experimental values of this "water" to "air" ratio, i.e., the ratio on the L.H.S. of Eq. (24), is satisfactory.

7. CONCLUSION

From the analysis given in Section 6, we see that the various theoretical predictions are borne out by the experiments.

Indeed, apart from the theory, our experimental result of Eq. (24) means that at $\mu r = 4.25$ in water the absorbed dose of a Ce(10/90) dosimeter is about 1.25 times that of a Ce(10/10) dosimeter ,that is:

$$\left[\frac{D(Ce(10/90))}{D(Ce(10/10))}\right]_{\mu r = 4.25} = 1.25 \quad (26)$$

as can be seen by repeated substitutions of Eqs. (20) and (21) for two chemical dosimeters into Eq. (24).

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DISCUSSION

H.H. EISENLOHR (*Scientific Secretary*): Did you take the real spectrum of the ^{60}Co source into consideration in your numerical calculations?

C.P. WANG: Yes, we did. Indeed, we considered the whole spectrum, line and continuum, in our numerical calculations of the build-up factors.

L. FITOUSSI: Your expression for the absorbed dose rate in Eq.(6), generalized by Eq.(17), is applicable not only under the conditions you set forth, but necessarily also under conditions of secondary charged particle equilibrium (the electrons in this case).

C.P. WANG: You are referring to photon-electron equilibrium. That is exactly what is assumed in our basic Eq.(1), from which Eq.(6) and Eq.(17), the latter generalized for an extended source, follow. Thank you for referring to this point.